Current and piezoresponse measurements of repolarized regions of thin $PbZr_{54}Ti_{46}O_3$ films

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Thin ferroelectric lead zirconate titanate films $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) are promising materials for piezoacoustic, capacitive, electromechanical and sensor devices [1]. PZT films have high dielectric permittivity and large piezoelectric coefficients when $x \approx 0.5$ (morphotropic phase boundary). In this work, the processes of local repolarization of these PZT films were studied by scanning probe microscopy (SPM) methods, in particular using the piezoresponse force microscopy (PFM) [2]. Also, the processes of current flow in repolarized regions were studied by conductive atomic force microscopy (CAFM).

Thin films of $PbZr_{54}Ti_{46}O_3$ were grown by high-frequency magnetron sputtering of a ceramic target with the addition of 10% lead oxide (PbO) [3]. The films were deposited at a temperature of 150° C for 40 minutes and 1 hour in an atmosphere of lead vapors. Then they were annealed at a temperature of 600°C. The growth was performed on a $Si/SiO_2/TiO_2/Pt$ substrate. The thickness of $PbZr_{54}Ti_{46}O_3$ films was d = 200 nm and 300 nm [4]. All SPM measurements were performed under room ambient conditions at the «Ntegra-Aura» and «Solver P47» (NTMDT) microscopes. In this work we used DCP01 probes (NTMDT) with a wear-resistant diamond coating.

In the investigated samples, the polarization of the PZT films was performed by applying a voltage to the film. Initially, the samples were not polarized, so it was possible to create polarized regions of different polarity (see Fig. 1). It was found that the repolarization occurs only when the value of the applied voltage U exceeds the value of the coercive voltage U_c for the samples under study. If the voltage U is applied to a small contact area with curvature (R ~ 20 nm), which is less than the thickness of the film under investigation (d> 200 nm), the resulting field in the film will be distributed unevenly. Near the boundary of the film with the substrate, the value of the field strength will be about $E \sim (2R/d^2)U$. The polarization of the region of the film under the probe will occur when the magnitude of this field exceeds the coercive field of the film $U>U_c= (d^2/2R)E_c$. The characteristic values of the coercive fields for thin PZT films are ~ 50-100 kV/cm [5], therefore for these films one can expect characteristic values of coercive voltages U_c ~ 5-10 V.

Figure 1 shows the PFM image and the current map I(x,y) of the surface of 200-nm thick film, which was polarized by voltages U_{dc} = -10V and U_{dc} = +10V (dark and light areas in Fig. 1a, respectively). The corresponding averaged cross-section profiles are also given at the Fig. 1. In the PFM image (Fig. 1a) regions with different polarity of residual polarization (P_r) are clearly seen. It should be noted that these results correlate with the results for a 300-nm thick film annealed at 545°C in [2]. In the «dark» region (Fig. 1a), the residual polarization vector ($-P_r$) is directed from the SPM probe to the substrate. In the «bright» areas (Fig. 1a), the residual polarization vector ($+P_r$) is directed from the substrate to the probe. In addition in Fig.1a (arrows) the boundaries of the «polycrystalline blocks» can be seen.

It should be noted that repolarized regions can also be found in the current map I(x,y) obtained with a small voltage at the probe U = +2V (Fig. 1b). A comparative analysis of the current maps and the distribution of the PFM signal indicate that the current is larger when the field and polarization directions coincide, and less when they are directed oppositely to each other [6]. Also, on the current map (Fig. 1b, arrows) of polarized regions, the boundaries of «polycrystalline blocks» are identified as regions of increased conductivity. It should be noted that in unpolarized films the boundaries of the blocks were not visible. A completely similar situation was observed also for a 300-nm thick film.

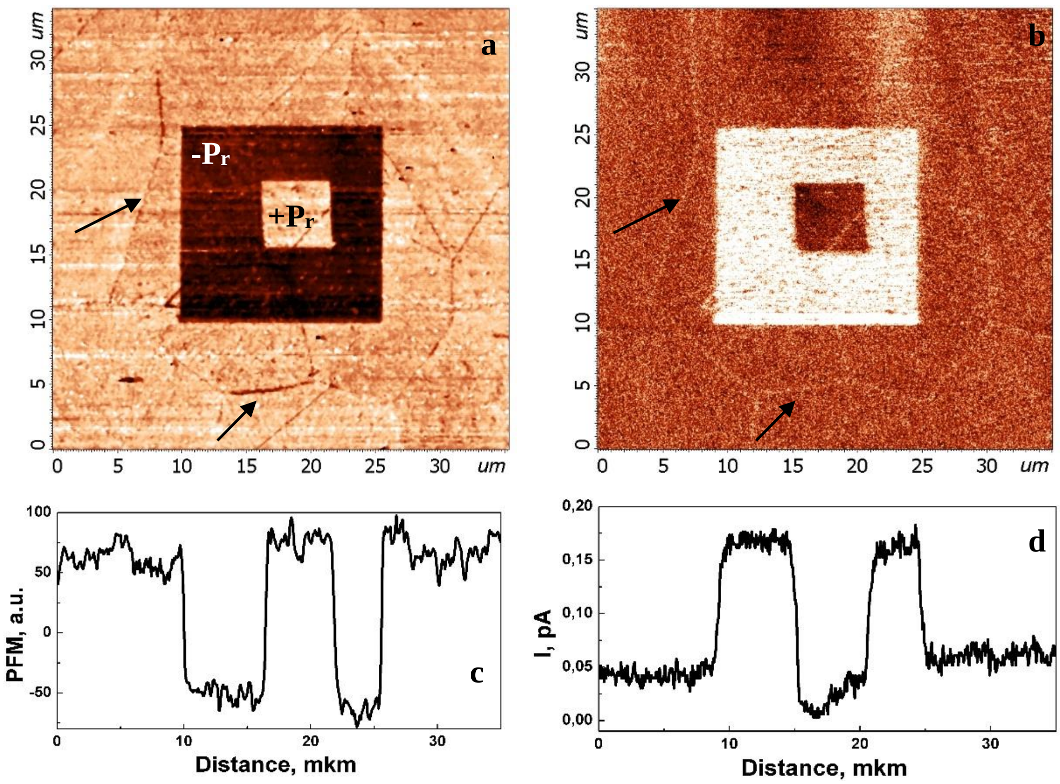

Figure 1. (a) PFM image of domains created when supplied to the probe U_{dc} = ±10V and (c) the corresponding averaged PFM profile; (b) the current map I(x,y) (at voltage +2V) of the same region and (d) the corresponding averaged current profile.

Thus, the technique of creating stable polarized regions (by applying 10V and more voltages) and their visualization in PFM and CAFM signals has been used for the study of piezoelectric and conductive properties of PZT films. The dependence of these signals on the thickness of the films is established. The dependence of the current on the polarization direction is established. It is also found that the polarization of regions allows revealing of the polycrystalline blocks boundaries as areas of increased conductivity.

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1. Scott J.F., *Science* **315,** 954-959 (2007).
2. В.В. Осипов, Д.А. Киселев и др., *ФТТ* **57**, 9, 1748 (2015).
3. И.П. Пронин, Е.Ю. Каптелов и др., *ФТТ* **52**, 124 (2010).
4. В. П. Пронин, А. Г. Канарейкин и др., *Поверхность*, **2** ,1 (2014).
5. J. Pérez de la Cruz, A. L. Kholkin, et.al., *J.Appl.Phys*. **108**, 114106 (2010).
6. Л.А. Делимова, Е.В. Гущина и др. *ФТТ* **56**, 12, 2366 (2014).